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Developing New 2D Materials and Heterostructures for Printed Digital Devices

2D-PRINTABLE - Deliverable report

D5.3. – Electrical characterization of films and vertical heterostructures

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Author(s)	Georg Duesberg (UniBw M) Kangho Lee (UniBw M)	2026-08-13
Checked by	Georg Duesberg (UniBw M)	2026-08-19
Reviewed by	Paolo Samori (UNISTRA) Zdenek Sofer (VSCHT)	2026-08-18
Approved by	Jonathan Coleman (TCD) - Project Coordinator	2026-08-19
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Project Scientific Abstract

The 2D-PRINTABLE project aims to integrate sustainable large-scale liquid exfoliation techniques with theoretical modelling to efficiently produce a wide range of new 2D materials (2DMs), including conducting, semiconducting, and insulating nanosheets. The focus includes developing the printing and liquid phase deposition methods required to fabricate networks and multicomponent heterostructures, featuring layer-by-layer assembly of nanometer-thick 2DMs into ordered multilayers. The goal is to optimize these printed networks and heterostructures for digital systems, unlocking new properties and functionalities. The project also seeks to demonstrate various printed digital devices, including proof-of-principle, first-time demonstration of all-printed, all-nanosheet, heterostack light-emitting diodes (LEDs). In conclusion, 2D-PRINTABLE will prove 2D materials to be an indispensable material class in the field of printed electronics, capable of producing far-beyond-state-of-the-art devices that can act as a platform for the next generation of printed digital applications.

Public summary

Deliverable D5.3 reports the electrical characterisation of printed two-dimensional-material films and selected vertical heterostructures developed in WP5. The work establishes quantitative links between deposition-controlled nanosheet assembly, microscopic junction transport and device-level electrical response.

For printed electrochemically exfoliated MoS_2 networks, impedance spectra from interdigitated-electrode test structures were analysed across channel lengths of 35-150 µm. Equivalent-circuit fitting separates nanosheet resistance, junction resistance, junction capacitance, contact resistance and network conductivity. The measurements show strong deposition-dependent differences: liquid–liquid interfacial deposition produces the lowest junction limitation and can approach a junction-to-nanosheet resistance ratio of about one. An impedance-derived network order parameter further connects electrical homogeneity with nanosheet alignment and film-thickness uniformity.

Hybrid COF-Azo/MoS_2 vertical heterostructures demonstrate how functional molecular layers can actively tune the electronic response of two-dimensional semiconductors. Integration of COF-Azo onto monolayer MoS_2 modifies the on-state current and threshold voltage, while reversible trans–cis isomerisation of the azobenzene units enables repeatable modulation of the electrical response under UV irradiation and thermal treatment. These results highlight molecular design, interface quality and electronic coupling as key parameters for introducing externally controllable functionality into hybrid 2D heterostructures.

Printed type-II ITO/PEDOT:PSS/Nb:WSe2/ZnO/PEIE/Al heterojunction diodes show clear dark-current rectification, with a rectification ratio of 26 at ±1 V. Together, these results provide practical electrical benchmarks for printed films and vertical stacks while identifying two remaining optimisation needs: Suppression of low-temperature measurement artefacts and reduction of shorting in fully nanosheet diode structures.

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Abbreviations & Definitions

Abbreviation	Explanation
2DM	2-dimensional material
C_J	Junction capacitance
COF-Azo	Azobenzene-functionalized covalent organic framework films
EE	Electrochemical exfoliation
FIB	Focused-ion beam
IDE	Interdigitated Electrode
ITO	Indium tin oxide
J	Current density
KPFM	Kelvin probe force microscopy
LLID	Liquid-liquid interface deposition
n	Network order parameter
PEDOT:PSS	Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate
PEIE	Polyethylenimine ethoxylated
R_C	Contract resistance
R_J	Inter-nanosheet junction resistance
R_{NS}	Nanosheet resistance
SEM	Scanning electron microscopy

1 Introduction

WP5 has focused on the electrical characterisation of printed 2DM films and heterostructures. This deliverables consolidate the directly relevant T5.2 and T5.4 results: Quantitative separation of nanosheet and junction transport in printed networks, and device-level characterisation of the printed dielectric layers, transistor and type-II semiconductor heterostacks.
A common-impedance characterisation platform now benchmarks deposition-dependent transport in printed MoS_2 networks, providing quantitative access to nanosheet and junction contributions and their relationship to network morphology. Hybrid COF-Azo/MoS_2 vertical heterostructures demonstrate the integration of functional molecular layers with 2D semiconductors and enable reversible modulation of the MoS_2 electrical response through photo- and thermally induced molecular switching. The printed Nb_2/ZnO diode provides an initial type-II heterojunction platform. Fully nanosheet diode stacks remain under optimisation because early all-nanosheet devices exhibited excessive shorting.

2 Methods and core part of the report

2.1 Printed network structure

EE MoS_2 nanosheet networks were deposited on pre-patterned lateral interdigitated electrode (IDE) arrays by liquid-liquid interface deposition (LLID), dip coating, spray coating and spin coating. The IDE channel length was varied from 30 to 350 µm so that fitted electrical parameters could be analysed as a function of device geometry and compared across deposition methods under a common protocol.

2.2 Impedance spectroscopy

Frequency-dependent impedance spectra were fitted with an equivalent Randles circuit comprising a series resistance and a parallel resistor-capacitor element. Channel-length-dependent analysis of the fitted series resistance, parallel resistance and capacitance yielded mean nanosheet resistance (R_{NS}), inter-nanosheet junction resistance (R_J), junction capacitance (C_J), contact resistance (R_C), network conductivity and the network order parameter (n). Here, $n = 1$ denotes an electrically homogeneous network as lower values represent broader junction property distributions.
In-operando KPFM provided an independent local measurement of gradual potential drops within nanosheets and sharp drops at inter-nanosheet junctions. Preliminary temperature-dependent impedance measurements were additionally performed from 300 to 80 K and analysed using Arrhenius behaviour to estimate thermally activated transport barriers.

2.3 Vertical heterostructure

Type-II diodes were fabricated by spin coating EE 7% Nb-doped WSe_2 with ZnO nanoparticles in the architecture ITO/PEDOT:PSS/Nb:WSe_2/ZnO/PEIE/Al. PEDOT:PSS provides an ohmic contact layer to p-type Nb:WSe_2, while n-type ZnO forms the heterojunction. Dark current-voltage measurements were used to assess rectification.
Vertical heterostructure engineering was also explored by integrating azobenzene-functionalized COF films with monolayer MoS_2. The resulting COF-Azo@MoS_2 heterostructure enabled reversible and optically tunable modulation of the MoS_2 transport characteristics, demonstrating how functional molecular layers can be used to tailor the electronic response across vertically integrated 2D interfaces. For further details on the hybrid architecture and its electrical characterisation, please refer to WP3.
A hybrid stack combining directly patterned TAC-WS_2 with solution-processed MoS_2 and NiO demonstrated the feasibility of integrating bottom-up and colloidal 2D materials, while also identifying layer conductivity and interfacial charge transport as key limitations. For further details on this heterostructure approach, please refer to WP3. Building on these findings, fully solution-processed heterostacks were developed using colloidal monolayer MoS_2, with particular emphasis on solvent orthogonality and the formation of continuous, pinhole-free layers. The resulting architecture enabled stable electroluminescence, mechanical flexibility, and extension to other 2D

materials and substrates, demonstrating the versatility of the heterostack platform. For further details on the heterostructure design and device implementation, please refer to WP6.

3 Results & Discussion

3.1 Printed MoS_2 networks and films

The impedance response varies systematically with channel length and is well described by the selected equivalent circuit. This enables microscopic transport quantities that were previously inaccessible in whole-network measurements to be extracted and compared across deposition routes.

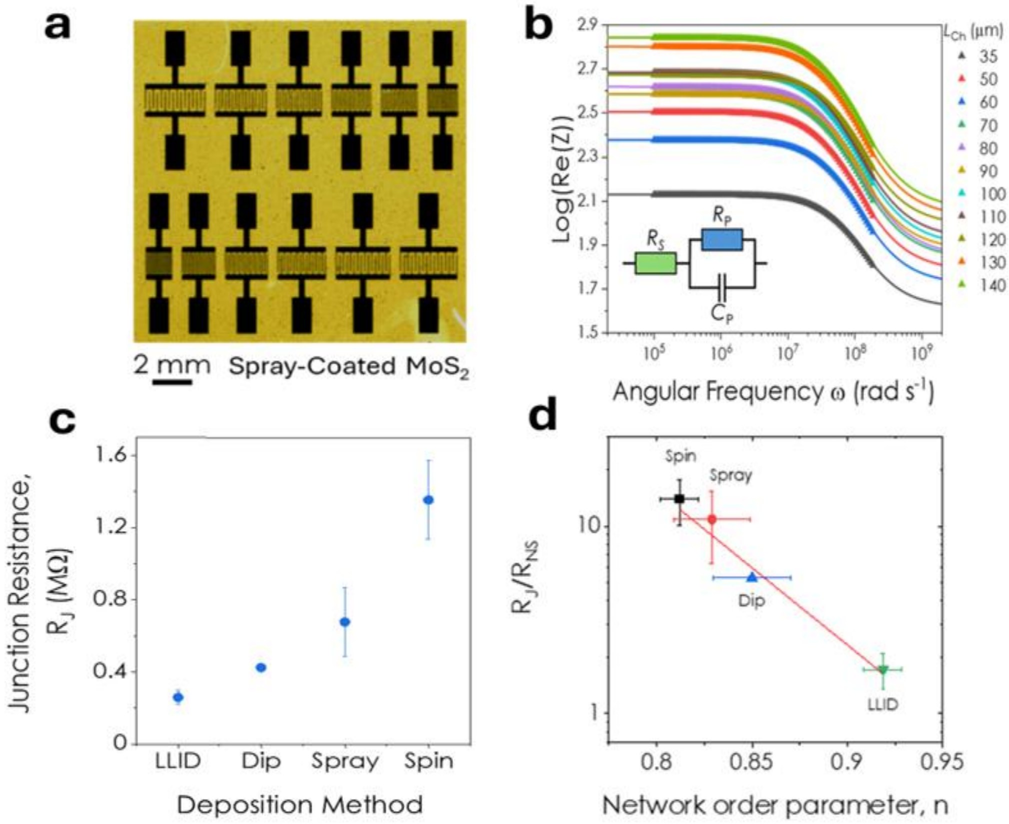

Figure 1. Electrical characterisation of printed EE MoS_2 nanosheet networks. **a)** IDE array with L_{Ch} = 35-150 µm. **b)** Representative Re(Z) spectra and equivalent circuit. **c)** Deposition-dependent junction resistance. **d)** R_J/R_{NS} versus the impedance-derived network order parameter *n*.

Junction resistance increases from LLID to dip-coated, spray-coated and spin-coated networks, showing that the deposition route controls inter-nanosheet overlap, conformality and junction quality (**Figure 1**). Because network conductivity and mobility depend strongly on R_J/R_{NS}, junction formation is a direct processing target. R_J/R_{NS} decreases strongly with increasing *n* and approaches approximately one for LLID films, linking improved electrical homogeneity to nanosheet alignment and film-thickness uniformity.

KPFM independently determined a mean R_J/R_{NS} of 10 ± 4 from local voltage drops, in good agreement with the impedance-derived values and supporting the network-scale model. Preliminary measurements from 300 to 80 K yielded similar activation energies of approximately 31-39 meV for LLID, spin-, spray- and dip-coated films. These temperature-dependent trends remain provisional because condensation artefacts were observed in the measurement setup.

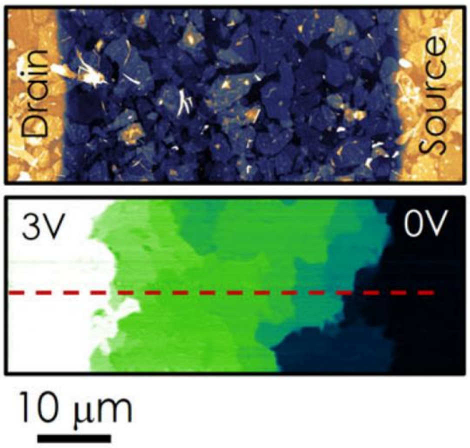

Figure 2. Topographic AFM image (top) and in-operando KPFM image (bottom) of a section of an EE MoS_2 network between source and drain electrodes.[1]

Spatially resolved KPFM maps further reveal that the potential varies relatively gradually within individual nanosheets, whereas pronounced voltage drops occur at nanosheet junctions, directly identifying the junctions as the dominant local resistive elements (**Figure 2**).

3.2 Printed type-II Nb:WSe_2/ZnO heterojunction diodes

The printed Nb:WSe_2/ZnO device exhibits clear dark-current rectification (**Figure 3**). The representative *J-V* curve presents a rectification ratio of 26 at ±1 V, confirming charge-selective transport across the p-type Nb:WSe_2/n-type ZnO interface.

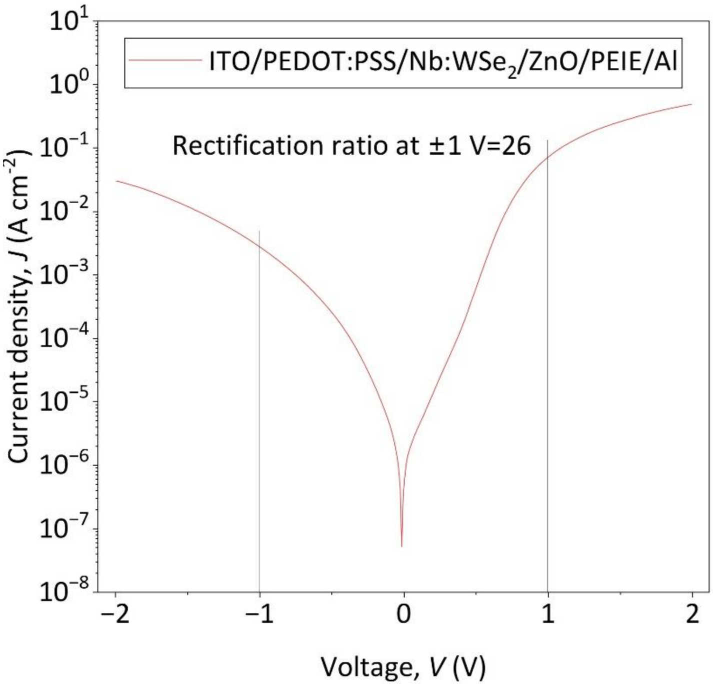

Figure 3. Dark *J-V* characteristic of the printed type-II ITO/PEDOT:PSS/Nb:WSe_2/ZnO/PEIE/Al heterojunction diode. The rectification ratio is 26 at ±1 V.

The present architecture uses conventional contact and ZnO layers because early all-nanosheet stacks shorted excessively. Replacement of ZnO with n-type EE WS_2 is ongoing and is the next step towards a fully nanosheet-based semiconductor heterostructure.

3.3 Hybrid COF-Azo/MoS_2 Vertical Heterostructures

Hybrid heterostructures combining functional molecular layers with 2D semiconductors were investigated as a route to tailor interfacial charge transport and introduce externally controllable functionality. COF-Azo films were integrated onto monolayer MoS_2 using an in-situ wet-transfer strategy, forming a vertically stacked COF-Azo@MoS_2 architecture. Integration of the COF layer substantially modified the MoS_2 transfer characteristics, including the on-state current and threshold voltage (**Figure 4**a, b), demonstrating that the functional overlayer effectively modifies the electronic response of the MoS_2 channel. Reversible trans–cis isomerisation of the azobenzene units under UV irradiation and thermal treatment further enabled repeatable modulation of the electrical response over multiple cycles (**Figure 4**c).

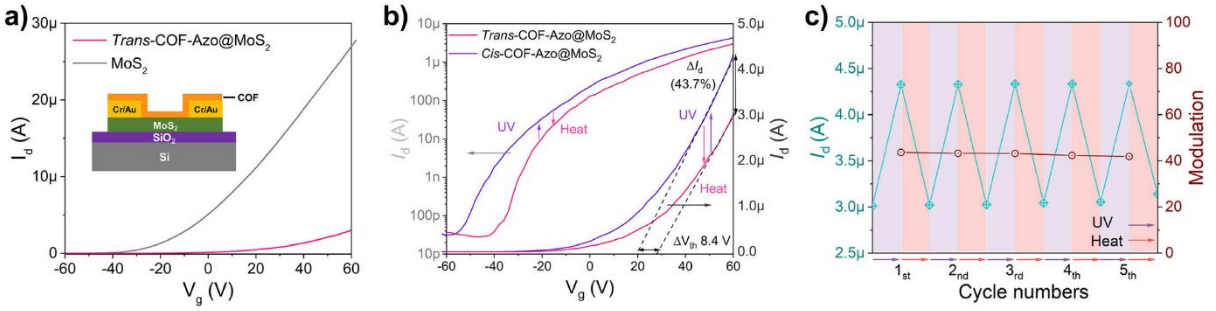

Figure 4. Photo-responsive behaviour of COF-Azo@MoS_2 heterostructures: **a)** Transfer characteristics before and after COF integration, **b)** UV-induced electrical modulation, and **c)** reversible switching over multiple cycles.[2]

These results demonstrate that vertical integration of functional molecular layers with 2D semiconductors can provide functionality beyond simple material stacking. In particular, the COF-Azo@MoS_2 system shows how molecular design and interface engineering can be used to actively tune charge transport in the underlying semiconductor. At the same time, the observed sensitivity of the electrical response to the interfacial molecular state highlights the importance of controlling interface quality and electronic coupling when developing more complex hybrid 2D heterostructures.

3.4 Contribution to project (linked) Objectives

The deliverable contributes directly to the WP5 objectives for the characterisation, understanding, and optimisation of charge transport in printed networks and heterostructures. In printed MoS_2 nanosheet networks, impedance spectroscopy combined with geometry-dependent analysis provides a systematic framework for separating nanosheet, junction, and contact contributions and relating them to deposition-dependent network homogeneity. Independent KPFM measurements further validate the dominant role of inter-nanosheet junctions, strengthening the link between processing, network structure, and electrical transport.

The work also extends from lateral nanosheet networks to vertically integrated material systems. Printed Nb:WSe_2/ZnO heterojunction diodes demonstrate charge-selective transport across solution-processed interfaces, while COF-Azo/MoS_2 and other hybrid heterostacks illustrate how vertical integration and interface engineering can introduce additional, externally controllable functionality. These activities support the WP5 objective of establishing structure–processing–transport relationships and translating them from printed networks towards functional heterostructures and devices.

3.5 Contribution to major project exploitable result

The results contribute to the project's exploitable outcomes by providing transferable characterisation methodologies and process guidelines for printed 2D material networks and heterostructures. In particular, the impedance-based approach offers a potentially reusable quality-control and process-optimisation framework that links deposition conditions to electrically relevant parameters such as junction resistance, contact resistance, and network homogeneity. Its agreement with local KPFM measurements further supports its use for benchmarking and comparing printed nanosheet networks.

Beyond network characterisation, the demonstrated heterostructures provide practical routes for evaluating and engineering charge transport across vertically integrated interfaces. The printed Nb:WSe_2/ZnO diode establishes a device-level platform for assessing heterojunction transport, while the COF-Azo/MoS_2 system demonstrates the integration of functional molecular layers with 2D semiconductors for tunable electronic behaviour. Together with the broader solution-processed heterostack activities, these results provide transferable know-how in electrical quality control, interface engineering, and scalable integration of 2D materials into functional electronic and optoelectronic architectures.

4 Conclusion and Recommendation

The results presented in this deliverable demonstrate a coherent pathway from network-level characterisation to the development and evaluation of functional heterostructures based on printed and solution-processed 2D materials. The impedance-based model provides an effective means of separating nanosheet, junction, and contact contributions within printed networks and relating these parameters to deposition-dependent network homogeneity. Independent KPFM measurements support this interpretation by directly identifying inter-nanosheet junctions as the dominant local resistive elements, providing a practical basis for systematic process optimisation and electrical quality control.

In parallel, the investigated vertical heterostructures demonstrate different routes towards integrating 2D materials into functional device architectures. Printed Nb:WSe_2/ZnO heterojunctions exhibit clear rectification, while hybrid COF-Azo/MoS_2 structures show that functional molecular layers can be integrated with 2D semiconductors to provide optically tunable electrical behaviour. Complementary heterostack activities further demonstrate the potential of combining bottom-up and solution-processed materials and highlight interface quality, layer conductivity, solvent compatibility, and film integrity as key factors governing successful vertical integration.

Based on these findings, further work should focus on three main directions: (i) validation of the impedance methodology across a broader range of materials, deposition parameters, and network architectures, including improved temperature-dependent measurements to clarify thermally activated transport; (ii) further development of fully nanosheet-based vertical heterojunctions, particularly through improved control of layer continuity, conductivity, and interfacial charge transport; and (iii) systematic investigation of functional hybrid interfaces to establish how molecular design, interface quality, and electronic coupling determine device-level transport and externally controllable functionality.

Particular emphasis should be placed on establishing reproducible process windows and quantitative performance criteria that enable direct comparison between different network and heterostructure configurations. The impedance methodology should also be further evaluated as a potential in-line or post-process quality-control approach for printed nanosheet networks.

Overall, the results provide both a transferable characterisation framework and practical strategies for integrating printed 2D materials into functional heterostructures. Further optimisation of junctions and vertically integrated interfaces is expected to improve their applicability to scalable printed-electronics and optoelectronic processes and to support the development of exploitable project outcomes within WP5.

5 Risks and interconnections

5.1 Risks/problems encountered

Risk No.	What is the risk	Probability of risk occurrence[1]	Effect of risk[1]	Solutions to overcome the risk
WP5.2	Network variability limits reliable transport parameter extraction	2	1	Standardised structures; statistical sampling; cross-validation)
WP5.3	Local measurements may not represent heterogeneous networks	3	2	Multiple-junction analysis; comparison with impedance
WP5.4	Non-uniform layers/interfaces cause leakage or poor charge transfer	2	1	Optimise layer quality, interfaces and processing

[1)] Probability risk will occur: 1 = high, 2 = medium, 3 = Low

5.2 Interconnections with other deliverables

The activities reported here are closely interconnected with developments in WP3 and WP6, linking materials and heterostructure development with electrical characterisation and device-level implementation. In relation to WP3, WP5 provides electrical characterisation of hybrid architectures developed from bottom-up and functional 2D materials. This includes the COF-Azo/MoS_2 heterostructure, where electrical measurements were used to assess the effect of the functional molecular overlayer on MoS_2 transport, as well as heterostack concepts combining directly patterned TAC-WS_2 with solution-processed 2D materials. These activities connect the material synthesis and integration approaches developed in WP3 with the investigation of interfacial charge transport and electronic functionality in WP5.

The link with WP6 is established through the development of solution-processed vertical heterostructures towards functional optoelectronic devices. The materials, interface considerations, and electrical transport concepts investigated within WP5 support the implementation of colloidal 2D semiconductors in multilayer device architectures, while WP6 extends these approaches to light-emitting devices, including flexible solution-processed 2D light-emitting devices. The activities across WP3, WP5, and WP6 establish a continuous pathway from material preparation and heterostructure assembly, through electrical and interfacial characterisation, to functional device integration.

6 Deviations from Annex 1

No deviations

7 References

1. Gabbett, C. *et al.* Understanding how junction resistances impact the conduction mechanism in nano-networks. *Nat. Commun.* **15**, 4517 (2024).
2. Wang, K. *et al.* Wet Transfer of In Situ Grown Azo-Containing Two-Dimensional Conjugated Covalent Organic Framework Films for Photoswitchable Electronic Devices. *Angew. Chemie Int. Ed.* **65**, (2026).